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Development of Laser Lift-off Process with a $GaN/Al_{0.7}Ga_{0.3}N$ Strained-Layer Superlattice for Vertical UVC LED Fabrication

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***Abstract*— A laser lift-off (LLO) process with a $GaN/Al_{0.7}Ga_{0.3}N$ strained-layer superlattice was newly developed for use in the fabrication of a vertical UVC LED without the use of UVC incompatible materials such as epoxy to suppress cracking. Since the UVC-LED epitaxial structures grown by Metal-Organic Vapor Phase Epitaxy contain AlGaN layers with high Al contents, it is often grown on an AlN buffer layer. In blue LEDs, GaN buffer layers are used for growth. However, GaN-based films often present a problem for UVC growth, resulting in cracking caused by lattice mismatch. AlN layers are transparent to UV lasers utilized in the LLO process and thus making lift-off of the sapphire substrate very challenging. This $GaN/Al_{0.7}Ga_{0.3}N$ strained layer superlattice was employed to absorb the UV laser during the LLO process and suppress the dislocations climbing to the UVC-LED epitaxial structure grown on this layer allowing for a highly uniform and crack-free surface. UVC-LED structures were grown utilizing a $GaN/Al_{0.7}Ga_{0.3}N$ strained layer superlattice inside a horizontal flow metal-organic vapor phase epitaxy reactor. Copper substrates were then deposited onto the back surface of the wafers. LLO was achieved by employing a laser fluence of 1 J/cm^2 from a 248 nm excimer laser through the sapphire substrate. Successful LLO of a 2" sapphire substrate was attained without any cracking introduced when using this process. No deterioration of crystal quality in UVC-LED epitaxial structure such as dislocations and intermixing of atoms by LLO was also confirmed by X-ray diffraction, scanning electron microscopy, and transmission electron microscopy analysis.**

***Index Terms*— $Al_XGa_{1-X}N$, Laser Lift-off, MOVPE, UVC**

I. INTRODUCTION

Recently, aluminum gallium nitride ($Al_XGa_{1-X}N$) based deep-ultraviolet (UVC) light-emitting diodes (LEDs) grown by Metal-Organic Vapor Phase Epitaxy (MOVPE) have garnered much attention. This interest attributed to their ability to emit at 200–280 nm and these wavelengths numerous applications in fields such as medical therapy, optical sensors, water sterilization, and disinfection. The high cost of bulk AlN substrates has resulted in most commercial UVC LEDs to utilize sapphire substrates for epitaxial growth [1-3]. These devices often suffer from current crowding and self-heating when using lateral chip processes for device fabrication. These issues often affect device efficiency, lifetime, and high current operation [4-5]. Performance of UVLEDs grown on sapphire and AlN substrates can be improved if both defects and resistivity are minimized.

UVC devices with a vertical LED structure can mitigate series resistance and current crowding issues by shortening the path between the n- and p- electrodes, which is often longer in lateral devices than vertical devices. The shorter path allows vertical UVC LEDs to have higher power saturation currents and longer lifetimes than those of lateral structures of the same epitaxial structure. Vertical LEDs also offer the possibility to improve light extraction compared with that in traditional lateral-based structures because another surface is exposed to which processes such as roughening, a technique proven to improve light extraction, can be applied [6]. In addition to the previously mentioned benefits, a vertical structure can allow bonding to thermally conductive substrates such as metals, which can lessen thermal-related issues related to current crowding and thermal impedance by acting as a large heat sink. A schematic illustration showing how lateral and vertical devices are often implemented are shown in Fig. 1 (a) and (b) respectively.

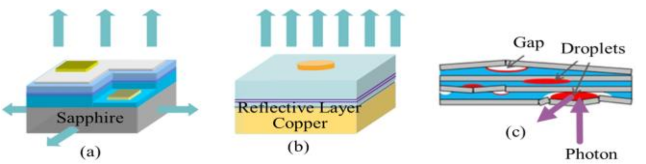

Fig. 1. Schematic structures of (a) a traditional lateral device, (b)typical substrate lifted vertical device and (c) SLS with Al droplets from post-LLO decomposition of low concentration $Al_XGa_{1-X}N$.[7)]

To fabrication vertical UVC LEDs, the substrate must first be removed from the device layers. There are numerous ways to remove the substrate, but one of the most proven techniques uses a UV laser to decompose specific layers of the epitaxial structure to enable lift off. This technique has been in use in the mass production of blue and ultraviolet (UVA) LEDs for over a decade. In this study, the focus is on using laser lift-off (LLO) technology to remove the substrate to realize a vertical UVC LED structure. This process is challenging, even for GaN-based devices in mass production, often the surfaces are damaged during LLO, and these issues are further compounded when high concentrations of Al is used with GaN due to mismatch induced cracking, and crystal degradation requiring novel techniques to overcome these challenges [6-10]. Combining low concentration Al content $Al_xGa_{1-x}N$ with GaN leads to complications due to the Al disassociation causing Al droplets to get caught between

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layers or surfaces. These Al droplets are hard to remove and stick very strongly causing damage as the two surfaces are separated. Schematic drawing of this type of situation is seen in Fig 1 (c). Takeuchi et al. and Adivarahan et al. have both noted that even when LLO is possible using low Al% $Al_xGa_{1-x}N$, the Al droplets left from the decomposition are sometimes a problem for lift off by inducing cracking from mechanical stress or blocking UVC light [7,11].

Unique challenges to achieve LLO for UVC-based devices is related to the transparency of the material to lasers in the UV range, requiring the inclusion of a sacrificial layer to facilitate LLO, which often negatively affects crystal quality. Such sacrificial layers are usually composed of GaN or $Al_xGa_{1-x}N$ with a low Al content that can absorb the laser energies. $Al_xGa_{1-x}N$ with a high Al content start to become transparent to the laser energy as Al percentage increases; making LLO not possible. Several groups have attempted LLO on DUV material systems with varying results requiring additional process and structures; such as extra strain management layers or patterned substrates, or epoxies that are not UVC compatible due to decomposition and yellowing. None of these groups were able to successfully emit photons below 280nm with their structures in a vertical configuration, which is in the UVC range used for sterilization [4,6-16]. The structure employed in this research is different from those of other groups mentioned: we attempt to insert a strained layer comprised of GaN and $Al_{0.7}Ga_{0.3}N$ and use the decomposition of the GaN layers for LLO. Note that the GaN layer absorbs a UV laser and $Al_{0.7}Ga_{0.3}N$ is transparent to a UV laser. The AlGaN layer with a high Al content was used to decrease the lattice mismatch between the SLS and a top $Al_{0.7}Ga_{0.3}N$ layer of the LLO template. We avoid the use of epoxy, which many groups depended on, which yellows and breaks down when exposed to UVC light. An example of such a device that depends on epoxy for successful LLO as a supporting material can be found in the literature [7,11,13,14,16]. Furthermore, to avoid any crystal degradation, the amount of $Al_xGa_{1-x}N$ with a low Al concentration or GaN needs to be minimized. It also needs to be easily removed because GaN and low concentration Al $Al_xGa_{1-x}N$ can absorb UVC light. Most groups have avoided using GaN in UVC LEDs because of its crystal degradation issues, but we show that this is not a problem if GaN/$Al_{0.7}Ga_{0.3}N$ strain layers are used. Likewise, aluminum droplets will not form from the disassociation of the $Al_xGa_{1-x}N$ because of the transparency of the $Al_{0.7}Ga_{0.3}N$. The lack of disassociation of the $Al_xGa_{1-x}N$ is important because aluminum droplets are problematic due to blocking of UVC light from escaping, and difficult to remove without damaging the surface of the LED as can be seen in Fig. 1(c). In this work, the LLO process takes advantage of a disassociation reaction of GaN that is facilitated by UV laser energy. This process has been commercially used for quite some time to produce blue and UVA LEDs. The chemical equation for this reaction is as follows:

$$GaN \rightarrow Ga + \tfrac{1}{2}N_2(g). \qquad (1)$$

This reaction leaves droplets of Ga or gallium oxide on the device surface, which then can be easily removed by etching using chemicals such as HCl [9]. LLO is inherently more difficult for UVC than UVB and UVA devices, because GaN not present and is often avoided due to absorption of the UVC spectra [7,11]. However, with LLO it is possible to remove the remaining GaN and SLS structures post process so that absorption is not a factor.

II. Experimental Work

A. LLO Structure Growth

The structure was grown by MOVPE using a horizontal flow under low pressure (10.5–14 kPa) at 1200°C. Precursor gasses used for MOVPE growth were trimethylaluminum, trimethylgallium, ammonia (NH_3), silane (SiH_4), and bis(cyclopentadienyl)magnesium (Cp_2Mg). SiH4 and Cp2Mg are used as precursors for n- and p-type doping, respectively. Sapphire (0001) substrates polished on both sides were heated to around 1200°C (set point) during growth.

Two original structures were grown for the development of this technology. The LLO template structure was composed of an undoped AlN layer (~250 nm thick), an undoped $Al_{0.7}Ga_{0.3}N$ layer (~300 nm thick), twenty pairs of GaN/$Al_{0.7}Ga_{0.3}N$ SLS (2 and 5 nm thick, respectively), and an undoped $Al_{0.7}Ga_{0.3}N$ layer (~300 nm thick). This structure was primarily used for developing the SLS stack. The other structure was a UVC LED composed of an undoped AlN layer (~250 nm thick), N-doped $Al_{0.7}Ga_{0.3}N$ (~650 nm thick), twenty pairs of GaN/$Al_{0.7}Ga_{0.3}N$ SLS (2 and 5 nm thick, respectively), and N-doped $Al_{0.7}Ga_{0.3}N$(~650 nm thick). Then, five pairs of $Al_{0.55}Ga_{0.45}N/Al_{0.70}Ga_{0.30}N$ multiple quantum wells, followed by a P-doped $Al_{0.80}Ga_{0.2}N$ electron blocking layer, and a P-contact and capping layer of P-doped $Al_{0.02}Ga_{0.98}N$ totaling approximately 1 µm thick were deposited on the structure. Schematics of the LLO template and UVC LED structures are presented in Fig. 2(a) and (b), respectively.

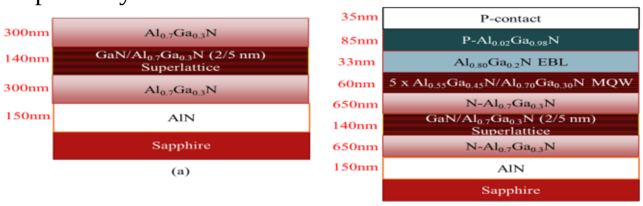

Fig. 2. Schematic cross-sectional structures of (a) lift-off template structure and (b)UVC-LED structure with thicknesses shown.

B. LLO Processing and Characterization

After epitaxial growth, wafers were inspected to check surface morphology. The LLO template and UVC LED epitaxial structure wafer types were then scanned by X-ray diffraction (XRD) and prepared for LLO processing by bonding to a copper substrate (150 µm thick). The laser dosage was 1 J/cm^2 from a KrF 248-nm laser through the sapphire substrate to treat the wafer, similar to other studies [4, 6-9, 11-16]. LLO templates were primarily used as a test platform for LLO processing. Fig. 3 outlines the simple LLO process using the LLO template structure from Fig. 2(a). In this process, the SLS layers are exposed to the laser through

the sapphire. The sapphire and layers before the SLS can then be removed, and the remaining structures can be left in place. In the case of UVC LED fabrication, the UVC LED structure would be grown on top of the template, bonded to copper substrates, and undergo LLO processing to remove the sapphire and buffer films, as we will demonstrate.

XRD was taken using a PANalytical X-Pert Pro system to characterize all films in both the symmetric (0002) and asymmetric (10-12) reflections. The thickness measurement and its mapping for a whole wafer were taken using Nanometrics RPM2000 equipped with reflectivity measurement system and a laser. The High-Angle Annular Dark Field Scanning Transmission Electron Microscopy (HAADF STEM) images were taken using Hitachi High Technologies HD-2700 at 200kV while the dislocation analysis by weak beam dark field images was taken using Hitachi High Technologies H-9000NAR at 300kV.

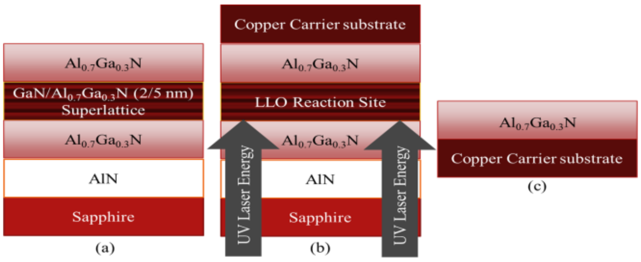

Fig. 3. Schematic simplified LLO process flow using LLO template structure.

III. Results and Discussion

A. *Results*

The quality of the crystal structure grown will ultimately determine if a technology is viable for production and worth investigating. Other groups have attempted similar lift-off systems with varying levels of success or processing complexity difficulties, but a structure using GaN in an SLS with a high concentration of Al ($Al_{0.7}Ga_{0.3}N$) has not been investigated [4, 6-9, 11-16]. The UVC LED structure utilized in this experiment appeared to have good surface morphology at 200× magnification, as seen in the image in Fig. 4(a) obtained from a Normarski microscope. The surface of the wafer is smooth and mirror-like indicating that the surface quality is high. Fig. 4(b) shows that wafer surface is regular and transparent. This transparency most likely achieved because the GaN layers are very thin and sandwiched between $Al_{0.7}Ga_{0.3}N$ layers, preserving material quality and preventing the strain-induced cracking usually seen in thick GaN layers combined with layers with high Al content [6-8, 10, 17-20].

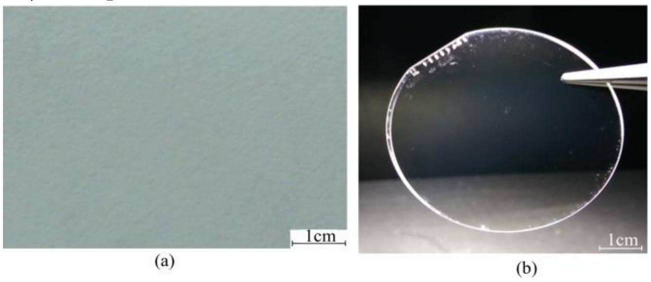

Fig. 4. (a) Micrograph of the wafer surface at 200x magnification and (b) typical wafer immediately post epitaxy.

B. *Effect of SLS Insertion on Crystal Quality*

The introduction of the SLS into a structure composed of an undoped AlN layer, undoped $Al_{0.7}Ga_{0.3}N$, 20 pairs of GaN/$Al_{0.7}Ga_{0.3}N$ layers as the SLS, and undoped $Al_{0.7}Ga_{0.3}N$ layer had a minor effect on crystal quality. The addition of the SLS increased the full width at half maximum (FWHM) of the (0002) diffraction from 109 to 224 arcsec and the FWHM of the (10-12) diffraction from 1108 to 1162 arcsec compared with those of pure AlN with an $Al_{0.7}Ga_{0.3}N$ top layer. These (0002) and (10-12) FWHM values are acceptable when compared with other values found in the literature for templates, and have been proven to be acceptable for the growth of deep UV and UVC LEDs, although it would be better to have a narrower FWHM for the (10-12) peak [2, 6, 8, 10, 17, 21-24]. It is possible that the large FWHMs were related to the thin layer thickness of the LLO template wafer (approximately 890nm thick). Fig. 5 shows the 2θ-ω scan of the (0002) reflection profile of the LLO template structure, which contains satellite peaks, indicating good crystal quality and abrupt interfaces.

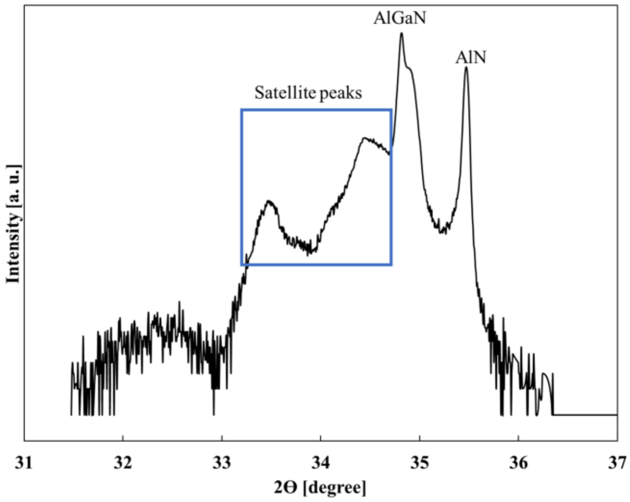

Fig. 5. 2Θ-ω scan of the (0002) reflection profile of LLO template structure.

This scan also indicates the presence of the SLS layers. The SLS layers are vital for lift off of this epitaxial structure because they enable it to absorb UV energy from the laser to induce the lift-off process. High-quality crystals are also essential for the growth of subsequent epitaxial layers of the UVC LED structure grown on top of the template structure.

C. *Post-Lift-off Analysis*

After copper bonding, the structure was exposed to a 248-nm KrF excimer laser through the sapphire substrate using a 1 J/cm^2 at 38ns pulse width dosage. Fig. 6 (a) shows the wafer immediately after LLO processing, clearly displaying a mirror-like finish as well as a residue on the sapphire. In this image, the left side wafer is metal side, and the right side is sapphire side after separation. Examination of the sapphire and metal wafer in SEM revealed that an epitaxial film with an approximate thickness of 0.77 µm remained on the sapphire side and a layer with a thickness of 1.03 µm stayed on the metal side. Micrographs of these films are presented in Fig. 6 (b) and (c), respectively with their schematic structures inset. Fig. 6 (d) depicts the thickness measurement taken before processing, which shows that the total epitaxial film

thickness is about 1.77 μm with excellent uniformity (4.23%).

Given the position of the SLS in the layers, these data suggest that the separation occurred at the SLS. The position of the separation indicates that the thin GaN layers were of appropriate thickness and composition to absorb the UV laser energy to allow LLO processing, which is later confirmed by TEM. TEM also revealed that only a single SLS pair was used for LLO, indicating that fewer pairs could be used without risking process safety.

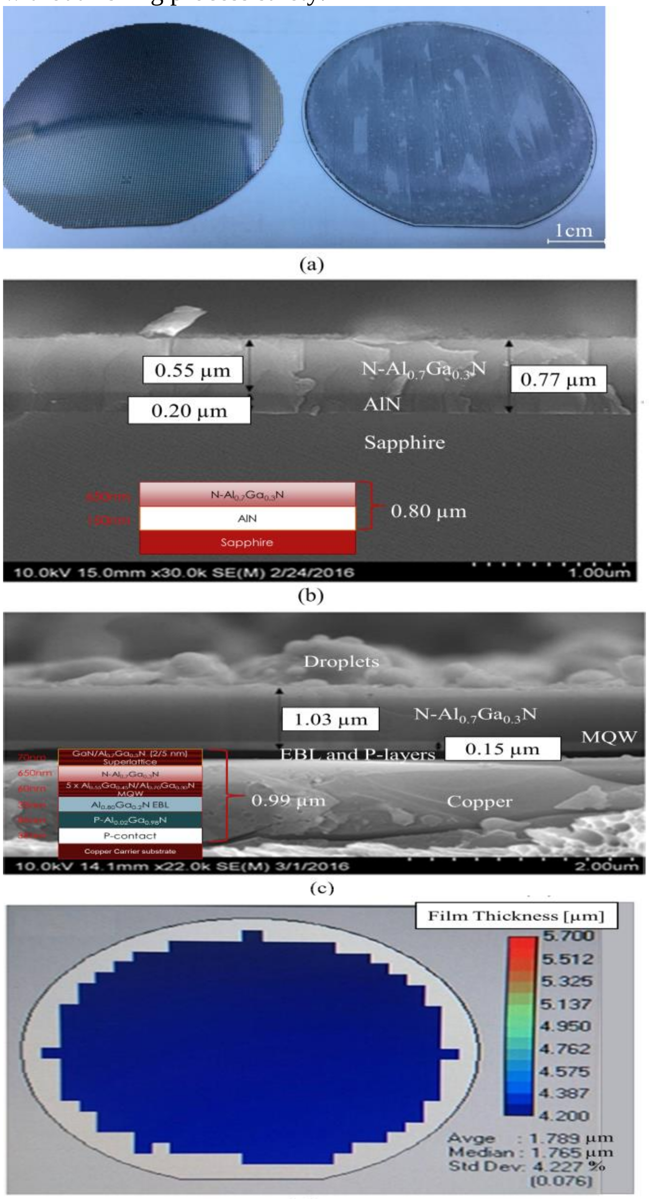

Fig. 6. (Color Online) Images of (a)the metal and sapphire wafers, (b)SEM of the sapphire wafer, and (c) SEM of metal-side wafer immediately post-LLO processing. (d) thickness measurement used for confirmation of film thickness.

After LLO processing, the wafer was cleaned with HCl to remove any remaining metal droplets and reveal the underlying surface morphology. Micrographs of the device at the n-face of the vertical device where LLO took place pre- and post-LLO treatment cleaning are presented in Fig. 7 (a) and (b), respectively. Fig. 7 (a) shows the surface with gallium metal droplets and the dark features are n-$Al_{0.7}Ga_{0.3}N$ post HCl cleaning, Fig. 7 (b) shows the same sample with gallium droplets removed revealing a smooth n-$Al_{0.7}Ga_{0.3}N$ surface remaining intact and smooth compared with the wafer surface before treatment. Furthermore, no obvious cracks and fissures were observed on the treated surface. Insets in these Fig. are schematic draws of a cross-section of the sample. Fig. 7 (c) and (d) show the difference between a good LLO surface and a cracked device induced by LLO; the cracked surface is easily observed under low magnification. Approximate percentage of cracked devices per wafer was <.001% with no special treatments or epoxy needed.

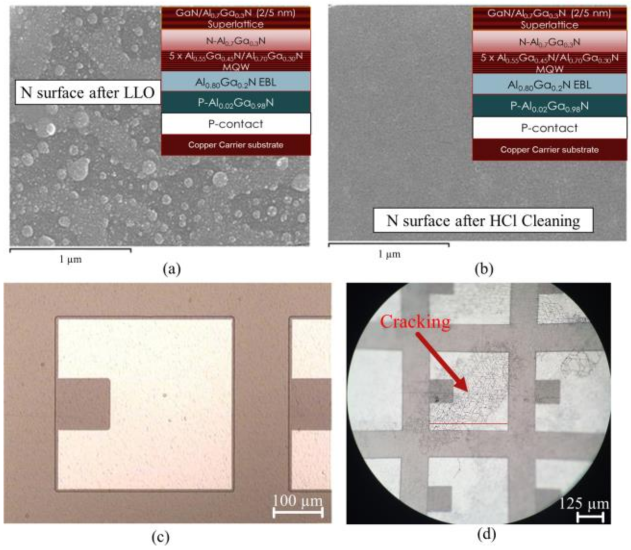

Fig. 7. SEM micrographs of the wafer surface (a) Pre-HCl cleaning the N surface post-LLO treatment and (b) post-HCl cleaning of the N surface post-LLO treatment with cross-section schematic inset. Examples of (c) successful LLO and (d) failed LLO.

The HAADF STEM images of the cross section of MQW and SLS in the post-LLO LED structure are shown in Fig. 8 (a) and (b), respectively. HAADF STEM imaging enables large contrast difference between the materials with different masses, allowing identification of layers by composition. In Figure 8 (a), bright narrower stripes correspond to $Al_{0.55}Ga_{0.45}N$ layers, while dark wider stripes correspond to $Al_{0.7}Ga_{0.3}N$ layers in MQW. In Fig. 8 (b), bright narrower stripes correspond to GaN layers, while dark wider stripes correspond to $Al_{0.7}Ga_{0.3}N$ layers in SLS. It is also confirmed that the designed structure is as expected based on our growth conditions of our MOVPE system. The HAADF STEM images also reveal that separation occurred using a single layer of the SLS system for LLO and that the surface and MQW structure remained intact and undamaged by the laser.

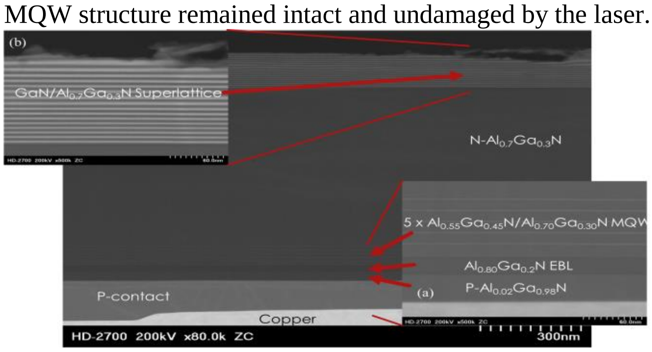

Fig. 8. The HAADF STEM images of the cross section of (a) active region and (b) SLS in the post-LLO LED structure.

The weak beam dark field images of the cross section of the post-LLO LED are shown in Fig. 9 (a) and (b). In Figure 9 (a) Burgers vector of (11-20) was used while that of (0002) was used in Fig. 9 (b). From this Burger vector analysis, line defects threading toward the growing direction observed in Fig. 9 (a) are not observed significantly in Fig. 9 (b). This result confirms that these line defects are threading dislocations. Similarly, a loop-shaped defect is observed in the center of Fig. 9 (b) while it cannot be observed in Fig. 9 (a). This result also confirms that this defect is a screw dislocation. From this, we can conclude that the LED and wafer are undamaged by the LLO and that this system enables LLO to work smoothly without structural damage. LED epitaxial structure was grown on the LLO template system to demonstrate that LLO could be safely conducted. In the TEM micrographs, it can be observed that the SLS layers are introducing no new dislocations. The SLS layers typically have the effect of bending dislocations and not introducing significant numbers of new ones simply by inserting them [10, 25-31]. By using thicker layers of $Al_{0.7}Ga_{0.3}N$ and keeping GaN as thin as possible in the GaN/$Al_{0.7}Ga_{0.3}N$ SLS, the overall lattice constant in the SLS layers approach a value nearly matching n-$Al_{0.7}Ga_{0.3}N$ minimizing the introduction of new defects due to critical thickness and lattice mismatch related issues. From TEM we can observe that most of the 20 SLS pairs that were inserted into the structure are intact, meaning that it can be concluded that it is safe to reduce the number of pairs for the LLO process, however, even with 20 SLS pairs, high quality, crack free surface was obtainable.

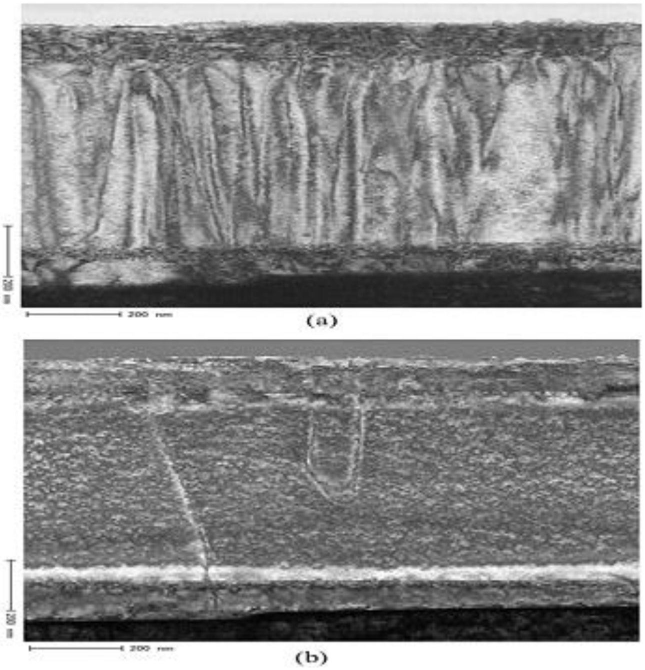

Fig. 9. The weak beam dark field images of cross section of the post LLO LED with (a) g=(11-20) and (b) g=(0002).

IV. Summary and Conclusions

Using XRD, the inserted GaN/$Al_{0.7}Ga_{0.3}N$ SLS system showed little impact on the quality of the crystal system, however, the introduction of this system allowed for the absorption of the laser to enable LLO processing to take place, due to the inclusion of GaN into the system. The $Al_{0.7}Ga_{0.3}N$ increased the layer's lattice constant enough to help bend and prevent the introduction of additional dislocation systems. Additionally, the $Al_{0.7}Ga_{0.3}N$ cannot be broken down by the laser, thus preventing Al droplets which make separation difficult.

SEM and TEM micrographs demonstrated that LLO indeed took place at the SLS, surprisingly, only one pair of SLS was sacrificed by this system for LLO to take place, meaning fewer than 20 SLS pairs could be used safely. Using this material system, it was possible to obtain clean, smooth surfaces after LLO without the introduction of epoxy or other special processing techniques to ensure a high quality lift off of the substrate system. Additionally, the removal of the substrate allows all traces of the SLS to be removed ensuring no GaN is left in the emission surface. Furthermore, this system is demonstrated to enable full 2" wafer level LLO instead of the chip or pixel LLO seen in the literature demonstrating that this technology can help enable the mass production of UVC vertical LEDs in the future. The ability to lift the substrates off AlN and $Al_XGa_{1-X}N$ materials can allow for a new wave of vertical based devices in other fields besides optoelectronics.

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